Neutron Symmetry and Gamma-Ray Measurements for the Fissile Material Transparency Technology Demonstration

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Introduction

The Fissile Material Transparency Technology Demonstration (FMTTD) at Los Alamos National Laboratory showed the determination of six plutonium attributes on unclassified plutonium objects and a U.S. nuclear weapon component. For the weapon component, all of the radiation data collected to measure the attributes were classified. The principal goal of the demonstration was to show that these measurements could be made using an information barrier that revealed none of the measurement data but presented useful results in the form of pass/fail indicators. The six attributes are:

- presence of plutonium
- presence of weapons-grade plutonium
- plutonium mass
- plutonium age
- absence of plutonium oxide
- axial symmetry of the plutonium source

The gamma-ray and symmetry measurements described here contributed wholly, or in part, to the determination of all six attributes. This paper expands upon the presentation, “Review of Plutonium Attribute Measurement Technologies: Gamma-Ray and Symmetry Measurements,” given during the actual demonstration on August 16, 2000.

Gamma-ray Hardware

The detectors used in the FMTTD comprised two high-purity germanium (HPGe) gamma-ray detectors and an improvised neutron multiplicity counter. All of the measurements were made of sources placed inside AL-R8 fissile-material storage containers inside the multiplicity counter. The gamma-ray spectrometers viewed the sources through the sides of the multiplicity counter (Figure 1).

Figure 1. The FMTTD detectors. In the center is the improvised neutron multiplicity counter. Flanking the multiplicity counter, in radio-frequency-shielded enclosures, are two high-purity germanium gamma-ray detectors.

As part of the layered architecture of the information barrier, we limited the collection of gamma-ray data to three narrow energy regions (Figure 2). This was accomplished by use of the upper- and lower-energy discriminators provided in the data-acquisition subsystems (two Canberra InSpectors™). Because each data-acquisition subsystem contains only one set of discriminators, three separate measurements were necessary. To keep the data-acquisition time reasonable, we employed two spectrometers.

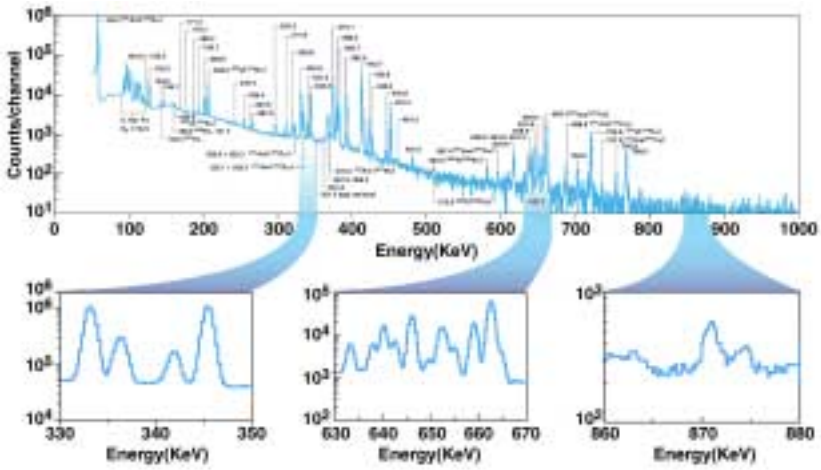

Figure 2. An illustration of the energy regions used for the gamma-ray measurements in the FMTTD. The upper spectrum from an unclassified U.S. weapons-grade plutonium metallic source illustrates the information richness of a plutonium gamma-ray spectrum. The lower spectra show expanded regions used for the FMTTD. The spectrum in the 900-keV region (far right) shows the presence of the 870.7-keV peak and was taken from a measurement of a PuO_2 source.

For simplicity, we refer to the three gamma-ray energy regions as the 300-, 600-, and 900-keV regions. These regions, of relatively high gamma-ray energy for plutonium isotopic and age measurements, were chosen because it was initially expected that plutonium sources of unknown geometry would be inspected. With the Russian Federation's decision to reshape its weapons-grade plutonium into standard shapes, this may no longer be necessary in an inspection regime. Whether or not other energy regions could be used depends critically on the geometry of the reshaped plutonium and its packaging. Nevertheless, the FMTTD was originally pursued on a tight time schedule, and this necessitated using some existing equipment and methods for the demonstration.

This resulted in the curious measurement geometry illustrated in Figure 1. In this geometry, the gamma-ray detectors view the AL-R8 storage container through four inches of high-density polyethylene located in the sides of the neutron multiplicity counter. In this case, the use of high-energy gamma-ray measurement regions was serendipitous, because only 25% of the primary gamma rays from our lowest energy region, the 300-keV region, reached the detector. In retrospect, the information barrier incorporated in the FMTTD measurement system worked so effectively that it was unnecessary to limit the measurements to these three narrow energy regions.

The hardware for the gamma-ray spectrometers comprised mostly commercial components. The detectors chosen were n-type to limit neutron damage in the germanium crystals. A detector of 50% relative efficiency was chosen for the measurements in the 300- and 600-keV regions. A detector of 100% relative efficiency was chosen to measure the more penetrating gamma ray at nearly 900 keV. This detector failed on two occasions, so a backup p-type detector of 67% relative efficiency was used in the demonstration.

The manufacturer of the HPGe detectors provides two signal leads. These signals were sent to two destinations: the first signal to an automated mechanical iris system and the second to the Canberra InSpector™. The iris comprises two leaves of machinable tungsten, nearly one-cm thick, which maintains the dead time in the detector at a relatively constant rate. This allows the use of a constant source–detector distance for sources of a wide dynamic range of emission intensities. The iris also has information barrier utility as it was implemented so that no information can be discerned about the strength of the source. A view of the detector, its iris, and the iris controller in the shielded enclosure is shown in Figure 3. A frontal view of one of the irises on a test stand is shown in Figure 4.

Figure 3. The HPGe detector used for the Pu-300 and Pu-600 methods. The detector is housed in a radio-frequency-shielded enclosure. To the right of the liquid nitrogen dewar is a heavy shield of machinable tungsten that surrounds the detector's cryostat. Above the shield is the stepper motor assembly that actuates the iris. To the right of the shield is an edge-on view of the autonomous tungsten aperture iris. In the upper left-hand corner of the enclosure is the circuitry that controls the iris.

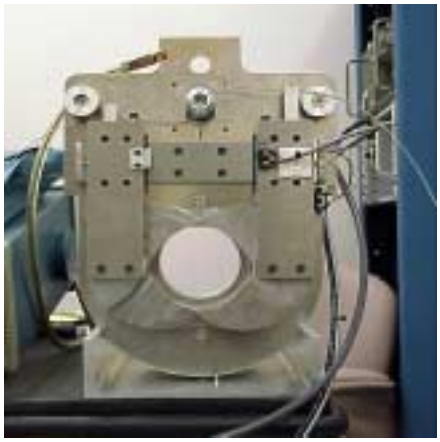

Figure 4. Frontal view of one of the autonomous tungsten irises.

The iris concept was originally formulated to deal with sources whose mass, geometry, and packaging vary widely in unknown ways. For the demonstration, the iris was designed to handle the widely varying input rates from the authentication sources. Of necessity, due to the original timeline for the demonstration, these sources were not specially prepared for the demonstration but assembled from materials on hand at LANL. In a final inspection regime involving plutonium reshaped to a single standard geometry, an aperture of fixed size might be employed. A fixed aperture is desirable, as it simplifies the measurement system and eliminates the maintenance associated with a moving aperture. Care is needed to prepare authentication sources

that can also be measured with the fixed aperture. It is possible, however, that a fixed aperture could have adverse information barrier implications. This possibility is discussed elsewhere.

The second signal from the HPGe detector is fed to a Canberra InSpector™ data-acquisition subsystem. This instrument combines a high-voltage supply, spectroscopy amplifier, and multichannel analyzer. The InSpector is controlled by a computer program written in the C programming language. The controlling computer is based on a simple PC104 single-board computer, assembled specifically for this application. The computer runs under the MSDOS operating system and takes advantage of the Canberra MSDOS libraries. The control program was written especially for the demonstration so that all unnecessary features of the commercial software provided with the InSpector would be eliminated. The single-board computer boots from a PROM and contains only volatile memory. In addition, to the MSDOS operating system, the PROM contains the control and analysis software for the system. When the system is actuated, the push of a single button initiates completely automatic operation. This eliminates any ambiguity in the operation of the system. These features implement various elements of the information barrier architecture, described elsewhere.

Gamma-ray Analyses

We developed several gamma-ray codes to determine wholly, or in part, five of the six attributes. These codes provide both the instrument control as well as the data analysis. There are three FMTTD codes: Pu-300, Pu-600, and Pu-900 corresponding to the acquisition and analysis of the data in the three energy regions of interest. Pu-300 determines the plutonium age—defined as the elapsed time since the chemical purification of the plutonium. This is accomplished by examining the ^{241}Am and ^{237}U ingrowth from the decay of the impurity isotope ^{241}Pu. Data in the energy region from 330–350 keV are used for this analysis. The Pu-600 method measures the ^{240}Pu/^{239}Pu ratio to determine if the plutonium sample is weapons quality. This analysis is carried out in the energy region from 630–670 keV. The presence of plutonium is determined by the presence of three characteristic ^{239}Pu peaks, one in the 300-keV region and two in the 600-keV region. The Pu-900 method determines the absence of PuO_2 by searching for a peak at 870.7 keV. If this peak is absent, then it is declared that there is no oxide in the sample. This peak arises from the de-excitation of the first excited state of ^{17}O.

Plutonium Age—The Pu-300 Method

The Pu-300 method determines the time elapsed since the last chemical separation of americium from the plutonium. The method exploits the fact that ^{241}Pu decays to ^{241}Am. Our measurement determines the ^{241}Am/^{241}Pu ratio. Because this ratio is a well-known function of time, the value of the ratio is a measure of the elapsed time since the chemical separation of americium. The measurement is slightly complicated by the fact that ^{241}Pu has a small α-decay branch to ^{237}U (see Figure 5 for simplified decay scheme). The β-decay branch to ^{241}Am accounts for 99.998% of the decays and the α-decay branch to ^{237}U accounts for the remaining 0.002% of the decays. Because the decay rates are known, as are the half-lives, the time since separation can be determined.

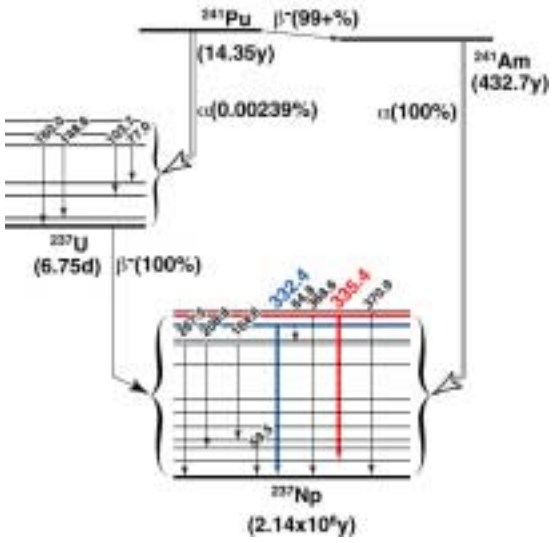

Figure 5. A simplified diagram showing the decay of ^{241}Pu showing the decay-mode branching and the 332.4- and 335.4-keV gamma-rays emitted from excited states of ^{237}Np that are the basis for the Pu-300 measurement technique.

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The Pu-300 method relies on the measurement of the 332.4- and 335.4-keV lines from excited states of ^{237}Np. While both ^{237}U and ^{241}Am populate these states, they are populated at different rates due to the differing decay branching of ^{241}Pu, the different specific activities of ^{237}U and ^{241}Am, and the greater likelihood of populating these relatively high-energy states due to the β-decay of ^{237}U than from the α-decay of ^{241}Am. An example of a portion of the pulse-height distribution used for analysis is shown in Figure 6. As can be seen from the figure, nature is sometimes unkind. The 332.4- and 335.4-keV peaks from ^{237}U/^{241}Am are tight doublets with ^{239}Pu peaks (at 332.8 and 336.1 keV) within less than one keV of the ^{237}U/^{241}Am peaks.

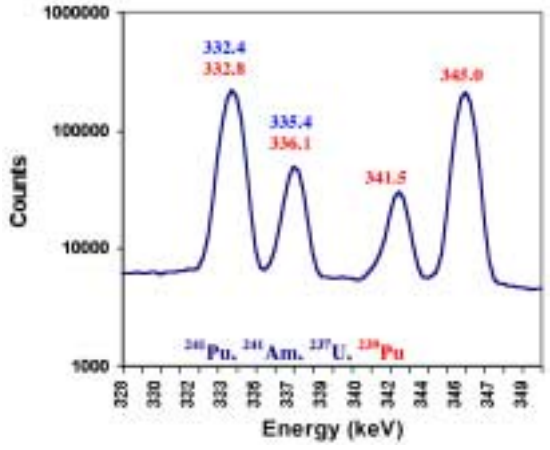

Figure 6. The portion of the gamma-ray spectrum that contains the key lines used in the Pu-300 method. The ^{237}U/^{241}Am peaks are indicated in blue, ^{239}Pu peaks in red.

Clearly, it is necessary to resolve the $^{237}U/^{241}Am$ peaks from the ^{239}Pu peaks. Fortunately, there are two nearby well-resolved single ^{239}Pu peaks at 341.5 and 345.0 keV. The peaks from ^{239}Pu at 341.5 and 345 keV constrain the amount of ^{239}Pu in the 332.4/332.8 and the 335.4/336.1-keV doublets. This essentially reduces the problem to solving two simultaneous equations with two unknowns to determine the $^{241}Am/^{241}Pu$ and thus determine the plutonium age.

For use in the FMTTD, a sample passed the age attribute if it was more than three years old.

Presence of Weapons-Quality Plutonium—The Pu-600 Method

In addition to age measurements, the same HPGe detector measures the isotopics of the sample. We call this measurement Pu-600 because it examines the gamma-ray pulse-height distribution in a region that spans, with nominal variation, 630–670 keV. The isotopic measurement actually determines the ratio of ^{240}Pu to ^{239}Pu. This measurement serves two purposes. The first is to determine that the plutonium is weapons-quality and the second is to facilitate the conversion of the effective ^{240}Pu value, measured by the neutron multiplicity counter, to total plutonium mass. Weapons-quality determination is made when the value of the $^{240}Pu/^{239}Pu$ ratio is below a threshold value of 0.1. During the automated analysis, the Pu-600 and Pu-300 measurements are done in series with the isotopic measurement performed first, followed by the age measurement.

The penetrating gamma rays in the 630–670 keV region ensure that measurements can be made regardless of the configuration of the plutonium item or its storage container. As with traditional high-resolution gamma-ray isotopic measurements used in international safeguards, analysis of the closely spaced peaks in a narrow region minimizes effects due to variation of detection efficiency and differential attenuation. Because we are determining the $^{240}Pu/^{239}Pu$ ratio from two closely spaced lines, we have no requirement to determine the absolute detection efficiency.

The analysis uses a variation of the safeguards gamma-ray analysis code, MGA. The procedure to determine the $^{240}Pu/^{239}Pu$ ratio is analogous to the determination of the age in Pu-300. The code fits the ^{239}Pu peak at 646 keV (Figure 7). The area in this peak and the emission intensities of the other ^{239}Pu lines in the region then constrain the peaks in the triplet in the region between 635–642 keV. When this triplet is symmetrical, as in Figure 7, the material is of weapons-quality.

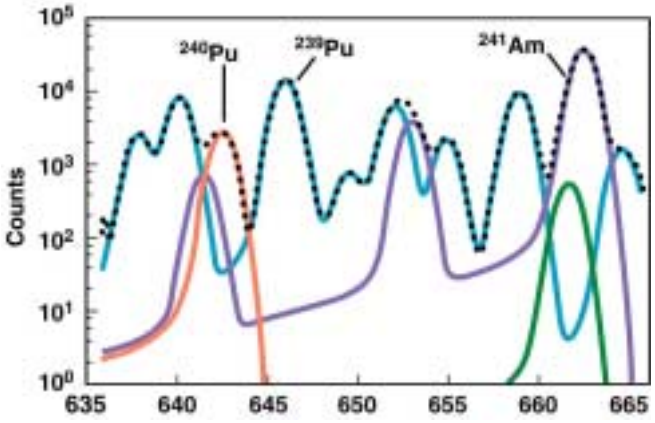

Figure 7. The region of the gamma-ray spectrum analyzed by the Pu-600 method. This plot shows the results of resolving the region into its isotopic constituents. The "dots" in the figure indicate the net measured data.

Some previous results obtained with this method from a variety of items are shown in Figure 8. The items were either "bare" or enclosed in AT400A or AL-R8 containers. The data in this figure show the Pu-600 isotopic ratios normalized to certified values of the ratios. Data acquisition was typically 15 minutes. Results plotted in green are from U.S. weapons-grade plutonium while results plotted in red are reactor-grade (e.g., the item designated PIDI-5 contained 22% ^{240}Pu). If the Pu-600 measurements were perfect, the results in the plot would be unity for all measurements. The average of these results is 0.99 ± 0.05.

We have been using and refining Pu-600 for six years. It has proven to be quite reliable and is the most mature gamma-ray technology shown in the demonstration.

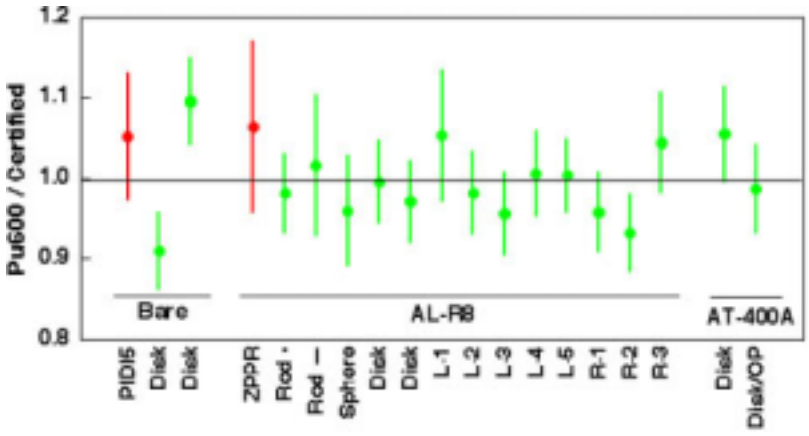

Figure 8. A variety of measured Pu-600 isotopic ratios normalized to certified values of the ratios. Results plotted in green are from U.S. weapons-grade plutonium while red results are reactor-grade. If the Pu-600 measurements were perfect, the results in the plot would be unity. The average of these results is 0.99 ± 0.05.

Plutonium presence—The Pu-300 and Pu-600 methods

In addition to the age and weapons-quality attributes, the Pu-300 and Pu-600 methods also determine the presence of plutonium. This is done by analyzing three characteristic photopeaks from ^{239}Pu. These include the 345 keV peak from Pu-300 and the 646- and 659-keV peaks from Pu-600. A declaration of plutonium presence requires that all three of these peaks must have areas that exceed five standard deviations of the underlying continuum.

Absence of plutonium oxide—The Pu-900 method

The newest attribute we have been concerned with is the absence of plutonium oxide. A more desirable attribute may have been determination of the presence of plutonium metal. However, in the short preparation time, the measurement community was unable to devise a technique for the direct determination of the presence of plutonium metal. If plutonium is present in a non-metallic form, it is likely to be present as plutonium oxide. We measured candidate oxide samples and found that an 870.7-keV line was present in all of the oxide samples and not present in the metal samples. This gamma ray arises from the decay of the first excited state of ^{17}O. We had initially thought that the mechanism for populating this state arose exclusively from ^{17}O(α,α'). However, it has become increasingly evident that this line can arise from the ^{14}N(α,p)^{17}O reaction. The interplay between these two mechanisms is unknown at this time and is the subject for research. Nevertheless, the presence of this peak *indicates* the absence of plutonium metal. The 870.7-keV peak was present in the plutonium oxide sample used as an authentication source in the FMTTD and absent in the metal samples.

For the FMTTD, we used the 870.7 photopeak to indicate the presence of plutonium oxide. If this peak was not present with an area exceeding five standard deviations above the background continuum, the absence of plutonium oxide was declared.

Neutron Symmetry Measurements

Under some circumstances, it may be important to know if the object in a storage container is cylindrically symmetrical, or not. We test for cylindrical symmetry by looking for an isotopic neutron radiation field emitted by a plutonium object in a sealed storage container. This method was suggested in Moscow by Vitaliy Dubinin of VNIIEF during discussions on the Mutual Reciprocal Inspections initiative in 1996. Shortly thereafter, the method was successfully tested in joint U.S.–Russian Federation experiments at Lawrence Livermore National Laboratory.

For these early experiments, neutron counts from unclassified plutonium objects in sealed storage containers were recorded following incremental rotations of 15º in front of fixed detectors (Figure 9). Ideally, if the item is cylindrically symmetrical, the neutron counts will be equal at each rotational position (Figure 10.). The 1996 experiments employed free-field measurements conducted on a low-scatter platform.

Figure 9. Dmitriy Semenov and Mikhail Chernov of RIPT (background) record neutron counts from their SRPS7 neutron detector (right) during 1996 joint experiments. LLNL technician Lori Switzer is rotating the AT400R container holding an unclassified plutonium object through an increment of 15º between measurements.

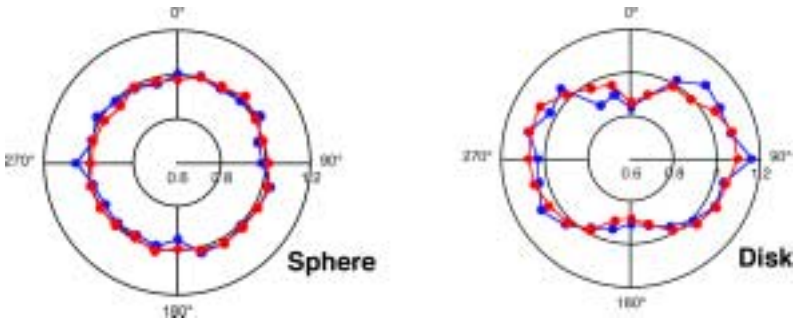

Figure 10. **(a) Polar plot of the neutron counts recorded from a plutonium sphere in an AT400R container during the 1996 joint experiments. The circular pattern indicates the presence of a cylindrically symmetric object in the container. (b) This polar plot produced from counts recorded from a thick plutonium disk shows that the neutron field is anisotropic, indicating asymmetry.**

For the FMTTD, a number of different plutonium attributes needed to be measured simultaneously. One of these attributes was the mass of the object in a storage container, measured with a neutron multiplicity counter. It was possible to conduct the mass and symmetry measurements simultaneously by tapping the neutron counts from the eight equally spaced neutron detector banks in the multiplicity counter (Figure 11). This arrangement satisfied the requirement of simultaneity. However, we feared that measuring this attribute in a moderated cavity, such as a neutron multiplicity counter, would considerably reduce the method's spatial resolution compared to the free-field measurements in 1996. This fear turned out to be unfounded, probably due to a layer of cadmium on the interior of the multiplicity counter.

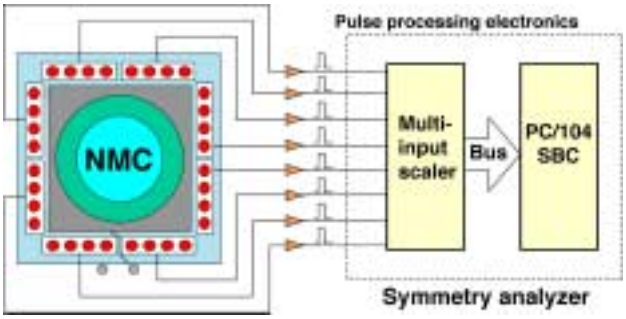

Figure 11. Block diagram of the neutron symmetry measurement made during the FMTTD. The eight outputs from the neutron multiplicity counter are recorded on an eight-input scaler and then analyzed by the PC-104 single-board computer.

Of course, another requirement for the FMTTD was that the measurements needed to be made behind an information barrier. As a consequence, data acquisition and analysis needed to be done by unattended computer control. Data from the eight scalers were automatically adjusted to reflect small efficiency differences in the eight detectors. The symmetry attribute was determined by a simple analysis of the adjusted net (background subtracted) detector counts to find the one detector that deviated the most from the average value, $\bar{y}$, of the adjusted net counts from all of the detectors. The absolute fractional deviation about the average, *s*, was computed.

$$s = \max\left(\frac{|y_i - \bar{y}|}{\bar{y}}\right), \quad \sigma_s \approx \frac{\sqrt{y_i}}{\bar{y}}$$

Where y_i = the number of net counts recorded in the *i*th detector bank,
$\bar{y}$ = the mean number of counts from all eight banks, and
σ_s = the standard deviation of *s*.

The values of *s* and s/σ_s were compared to an arbitrary threshold value chosen specifically for the FMTTD. To be declared asymmetric, the value of *s* had to exceed 0.15, and this value had to be statistically significant as determined by a value of s/σ_s in excess of 3.0.

Conclusion

We developed several gamma-ray methods to determine wholly, or in part, five of the six attributes. Codes provide both instrument control as well as data analysis. Three FMTTD codes—Pu-300, Pu-600, and Pu-900—correspond to the acquisition and analysis of the data in the three energy regions of interest. Pu-300 determines the plutonium age, defined as the elapsed time since the chemical purification of the plutonium. Pu-300 examines the ^{241}Am ingrowth from

the decay of the impurity isotope ^{241}Pu, using data in the energy region from 330–400 keV. The Pu-600 method determines the ^{240}Pu/^{239}Pu ratio to determine if the plutonium sample is weapons-grade or not. This analysis is carried out in the energy region from 630–670 keV. Pu-900 determines the absence of PuO_2 by searching for a peak at 870.7 keV. If this peak is absent, then it is declared that there is no oxide in the sample. This peak arises from the de-excitation of the first excited state of ^{17}O.

The neutron symmetry method examines the isotropy of the neutron emission field as measured by eight banks of detectors. This method is an extension of joint U.S.–Russian experiments conducted in 1996.

These methods have worked very well through many measurements. More work clearly needs to be done on the absence of oxide attribute and that is being pursued. All in all, we believe that we have four stable and very useful techniques for fissile material transparency.